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(54) Title: CRACKING FURNACE AND USE THEREOF IN THERMAL CONVERSION

(57) Abstract

Cracking furnace comprising a containment having a feed inlet and an outlet for cracked product, in which containment is arranged at least one radiant section comprising a single coil arranged around a radiant heat source in at least two co-axial rows, whereby the opening of the coil at that part of the coil forming the innermost row is connected to the feed inlet and the opening of the coil at that part of the coil forming the outermost row is connected to the outlet for the cracked product. Process for the thermal conversion of a hydrocarbon oil feed wherein use is made of the above cracking furnace.

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CRACKING FURNACE AND USE THEREOF IN THERMAL CONVERSION

The present invention relates to a cracking furnace and to thermal conversion processes wherein use is made of such furnace.

Cracking furnaces for thermal conversion operations are known in the art. A cracking furnace generally consists of a radiant section and an (optional) convection section. The radiant section consists of one or more radiation cells, each comprising one or more coils and a heat source, usually a burner. The word "coil", as used herein, has its common meaning of "a spiral pipe forming a continuous conduit", but it should be noted that in the art of thermal cracking, the said spiral pipe may have a secondary structure superimposed on its primary, circular shape. For example, the said pipe may have a series of axial (w.r.t. the coil axis) bends giving it a sinusoidal shape. It should be noted too that a "coil" may comprise a series of (concentric) rings or spirals, but also a single ring or spiral of such a series. The latter will also be referred to as "a single coil". The convection section may contain subsections for preheating the process feed, for superheating low pressure and medium pressure steam and/or for preheating the combustion air. Hot flue gases formed during combustion of the fuel in the burner in each radiation cell are passed from the radiation cell to the convection section where these hot gases are used to heat the coils in each of the aforementioned subsections.

Coke formation is the major factor determining the run length of a thermal conversion operation between two successive decoking/cleaning operations. Since coke formation is favoured by high temperatures during the

thermal conversion reactions, it is important to attain a low average heat flux inside the radiation cells. Conventional design average heat flux may range from about 10 kW/m^2 to about 30 kW/m^2. It will be appreciated that local coke deposit rates inside the coils are very important and are in fact the limiting factor with respect to run time. The peak heat flux, consequently, is a very important parameter. Therefore, number of coils, coil layout, dimensions of the radiation cell(s) and number and type of burners are normally selected to minimise the peak heat flux. In general, peak heat flux should not exceed about 75 kW/m^2 and temperature of the walls of the coils should not exceed about 625 °C in order to prevent local coke deposit rates becoming too high.

The radiant section of a conventional coil cracking furnace, particularly when used without subsequent soaker vessel, normally consists of two subsections: a heating section and a soaking section. Each of these sections may in return contain one or more radiation cells. In the heating section the feed is heated to such temperature that the cracking reactions are initiated and can proceed to a certain conversion level, whilst in the subsequent soaking section, the temperature is kept sufficiently high to allow the cracking reactions to proceed further in order to increase the conversion level. The piping used for the coils normally has the same diameter throughout the entire cracking furnace and this diameter is usually in the order of magnitude of about 4 inch (about 10 cm).

Although these coil cracking furnaces perform satisfactorily, there is still room for further improvement. Particularly in terms of conversion level, run time between two successive decoking operations, and costs involved in manufacturing and operating the

furnace, improvements are still possible. The present invention, therefore, aims to provide a cracking furnace which enables an increased conversion level when converting heavy hydrocarbon oil feeds at longer run times and reduced manufacturing and operating costs. More specifically, the present invention aims to provide a cracking furnace having a reduced sensitivity for coke formation, having an increased liquid residence time so as to increase the conversion level and having an integrated heating and soaking section. Furthermore, the present invention aims to provide a thermal conversion process, wherein such improved furnace can be used.

Accordingly, the present invention relates to a cracking furnace comprising a containment having a feed inlet and an outlet for cracked product, in which containment is arranged at least one radiant section comprising a coil and a radiant heat source, characterised in that a single coil is arranged around the radiant heat source in at least two coaxial rows, whereby the opening of the coil at that part of the coil forming the innermost row is connected to the feed inlet and the opening of the coil at that part of the coil forming the outermost row is connected to the outlet for the cracked product.

Any reference made to the geometry of the cracking furnace according to the present invention refers to the cracking furnace placed as it would be during normal operation.

Integration of the heating and soaking section, so that the heat generated by the radiant heat source is used more effectively, has been attained by applying one single coil which is arranged in the above defined manner around said heat source. By integrating the heating and soaking section, the total number of burners which form the radiant heat source required can be reduced, which

also reduces fuel consumption. It will be understood that this may cause significant savings on capital costs (fewer burners, fewer separate radiant sections and less measuring and control equipment) and operating costs (less fuel) as compared with the conventional furnaces having a separate heating and soaking section.

The innermost row of the coil, which is formed by that part of the coil being closest to the radiant heat source and hence has the highest peak heat flux, is equivalent to the heating section. Consequently, the feed inlet is connected to the opening of the coil at this innermost row, so that the cold or preheated feed can effectively be heated to the temperature at which the cracking reactions are initiated. The soaking section is formed by the outer row(s) of the coil and accordingly this section has a lower peak heat flux than the heating section formed by the most inner row. Because in the furnace according to the present invention no separate radiant heat source for the soaking section is applied, the peak heat flux of the soaking section in the present furnace is lower than that of the soaking section in a conventional furnace, so that coke formation is reduced and run time is increased. In a preferred embodiment of the present invention, the single coil is arranged in two rows around the radiant heat source. It has been found that in such two row-configuration, an optimum conversion level can be attained.

In order to increase the capacity two or more of the radiant sections or cells described hereinbefore may be combined in a larger containment. In such embodiment the inner rows of the coils of the individual radiant cells are suitably combined into a single inner row, whereby there is a common outer row surrounding the inner row. This is schematically illustrated in Figure 3.

The single coil can be made from a tube of any material known in the art to be suitably applied for furnace coils. Normally these materials are selected on the basis of sulphur concentration and total acid number of the feedstock to be treated, and coking resistance (ability to prevent or suppress coke deposition on the surface of the material at high temperatures). Suitable materials, for instance, are those based on alloys of chromium and molybdenum and on alloys comprising chromium and nickel as the major constituents. Beside these main metals, small amounts of other components like carbon, manganese and/or silicium may also be present. Trace amounts of elements such as niobium, titanium, tungsten and zirconium are sometimes present as well. A specific example of a suitable material is a chromium-molybdenum alloy, wherein the atomic ratio of chromium to molybdenum is between 8 and 12, more suitably between 8.5 and 10.5. Another example is an alloy comprising nickel and chromium as main components (Ni/Cr weight ratio of between 1.1 and 1.7, suitably between 1.2 and 1.5. A very specific example of the latter is an alloy comprising 33-38% by weight nickel, 23-28% by weight chromium and small amounts of carbon (0.35-0.60% by weight), manganese (1.0-1.5% by weight) and silicium (1.0-2.0% by weight) as well as a trace amount of niobium. The latter material is commercially available as MANAURITE 36X (trade mark).

The alloys comprising nickel and chromium as main components, especially the alloys described above, have a much higher coking resistance than the alloys based on chromium and molybdenum. Consequently, furnaces comprising a coil based on an alloy comprising nickel and chromium as main constituents can be operated up to higher coil skin temperatures without coke deposition onto the inner coil wall or skin occurring in such amounts that operation has to be stopped for decoking

than when using the alloys based on chromium and molybdenum. This, in return, allows longer operation times.

Although diameters of less than 4 inch (10 cm) may be used, the diameter of the coil (more precisely: the inner diameter of the pipe making up the coil) is suitably more than 4 inch (10 cm), i.e. more than the diameter normally used in conventional coil furnaces. Such increased diameter, namely, results in longer liquid residence times, at equal feed rate, and hence in a higher conversion, whilst the coil is also less prone to coke formation, particularly in situations where a pressure drop occurs. It has been found particularly advantageous to employ a coil of which the diameter at the feed inlet is smaller than the diameter at the outlet for the cracked product. Even more preferably, the diameter of the coil is the same throughout the part of the coil forming the innermost row. This implies that the expansion of the diameter occurs in that part of the coil forming the outer row(s).

In general, the diameters of the coils are determined on the basis of feedstock characteristics, such as volatility, cracking behaviour and two phase flow pattern. The diameters are selected such that liquid loading and/or flow pattern instability are permitted to a certain extent. Preferred diameters of the coil, then, at its inlet, i.e. at the start of the innermost row, are in the range of from 4.5 to 10 inch (11 to 25 cm), more preferably in the range of from 5 to 7 inch (13 to 18 cm). The diameter of the coil at the outlet for cracked products is suitably more than 4.5 inch (11 cm), preferably in the range of from 5 to 12 inch (13 to 30 cm) and even more preferably in the range of from 6 to 10 inch (15 to 25 cm).

The shape of the coil should be such that the feed can be heated to or kept at the desired temperature whilst coke build-up on the inside of the coil is suppressed as much as possible. Furthermore, the length of the coil in combination with the velocity of the feed through the coil should be such that the residence time of the feed is sufficiently long to allow the desired level of cracking to occur. It will be appreciated that optimum heat transfer of the radiation heat via the wall of the coil is very important. Accordingly, the surface of the coil's wall directed towards the radiant heat source should preferably be maximised in order to allow an optimum heat transfer. It has been found particularly advantageous to employ a coil which is made from a sinus-shaped tube arranged in a plane substantially perpendicular to the cross-section through the radiant heat source. Such shape, namely, allows long residence times and hence high conversion of a hydrocarbon oil feed, but also allows a favourable flow regime and optimum surface exposed to the radiation heat, so that an optimum heat transfer via the walls of the tube can be achieved.

The radiant heat source suitably comprises at least one burner and is suitably arranged in the middle of the bottom of the radiant section and, accordingly, in the centre of the coil. For reasons of process control, the number of burners will usually be kept limited. In practice, this implies that normally not more than four burners are used in a single radiant section. Any type of burner known to be applicable in cracking furnaces can in principle be applied. Examples of suitable burners, then, include steam-atomised oil burners, pressure-jet-atomised oil burners and gas burners. Examples of steam-atomised burners are the Lyunet burner and the Pillard burner (Lyunet and Pillard are trade marks). Preferably, the radiant heat source comprises three to six burners,

whilst in a highly preferred embodiment four burners are used as the radiant heat source.

The walls of the containment of the present cracking furnace are suitably made of a ceramic material, which is the material normally applied in the art. The inner diameter of the containment (hereinafter called "the inner lining") may suitably be within the range of from 4 to 15 m, but more suitably has a value of from 8 to 10 m. The thickness of the ceramic wall is largely determined by the accepted heat loss (usually 1 to 3%) and the thermal conductivity of the ceramic insulating material itself. Accordingly, the ceramic wall may have a thickness of from 10 to 40 cm, suitably of from 20 to 40 cm.

The cracking furnace according to the present invention may be a horizontal or a vertical furnace, the latter being preferred, because it requires less area in a refinery and because the flow regime in vertical tubes is in general more favourable, especially at reduced throughputs. The cross-section of the radiant section of the cracking furnace may be circular, square or rectangular. For the purpose of the present invention a vertical furnace with a radiant section having a substantially square horizontal cross-section, whereby the single coil is arranged in two substantially square rows around the radiant heat source is preferred. A square shape of the radiant section offers additional advantages with respect to the circulation of the flue or combustion gases formed during operation of the burners. In a vertical furnace such square shape, namely, causes the flue or combustion gases formed to flow down via the corners of the radiant section which in return allows the burners to have a stable flame pattern.

The single coil cracking furnace according to the present invention offers an additional advantage with

respect to decoking. At the end of a run time, when the pressure drop caused by plugging of the coil with coke has reached its maximum allowable value, the operation is stopped and the coil must be decoked. Several methods for decoking are known in the art and a particularly suitable decoking method for coil crackers is steam/air decoking. In this method, steam is introduced into the coil and the burners are set at a lower flame than during the cracking operation. Air is subsequently added to the steam in such quantity that the cokes are burnt (incompletely) and gasified to form a gas mixture of carbon monoxide and hydrogen, which is removed from the coil together with other impurities deposited inside the coil during the cracking operation. The fact that the furnace according to the present invention comprises a single coil only considerably facilitates such decoking operation.

In addition to the radiant section(s), the cracking furnace according to the present invention may have at least one convection section arranged therein. As has been explained above, the convection section may contain subsections for preheating the process feed, for superheating low pressure and medium pressure steam and/or for preheating the combustion air. The hot combustion gases formed during combustion of the fuel in the burner are passed from the radiant section to the convection section where these hot gases are used to heat the coils in each of the subsections. For an optimum heat balance of the furnace, it is preferred to include at least one convection section.

A most preferred cracking furnace, then, is a vertical cracking furnace with the radiant section having a substantially square horizontal cross-section, wherein the single coil is a sinusoid-shaped tube arranged in a plane substantially perpendicular to said cross-section, thereby forming two substantially square rows around the

radiant heat source, and wherein the diameter of the coil is the same throughout the inner row having a value in the range of from 5 to 7 inch (13 to 18 cm) and expands in the outer row to a value in the range of from 7 to 9 inch (18 to 23 cm).

The present invention also relates to a process for the thermal conversion of a hydrocarbon oil feed wherein use is made of a cracking furnace as described above.

Thermal conversion processes involving the use of coil cracking furnaces are well known in the art. In general, the main requirement to be met by the furnace employed in such a process is to heat the feedstock to a sufficiently high temperature and keep it at that temperature for a sufficiently long time to obtain the desired cracking severity or conversion. Typically, operating temperatures are in the range of from 350 to 650 °C. For the purpose of the present invention, operating conditions preferably are such that a 520 °C+ conversion of at least 35%, preferably from 45 to 90%, more preferably from 60 to 85%, is attained. The expression "520 °C+ conversion" as used in this connection is defined as the weight percentage of material present in the feed having a boiling point of 520 °C or higher, which is converted into material having a boiling point below 520 °C.

The hydrocarbon oil feed to be converted suitably is a hydrocarbon oil fraction comprising substantial amounts of material having a boiling point of 520 °C or higher. Examples of suitable feedstocks are atmospheric residues (long residues), vacuum residues (short residues), deasphalted residual oils or mixtures of two or more of these. Deasphalted residual oils are preferred feedstocks.

In the present process, the hydrocarbon oil feed is introduced at the feed inlet and enters the coil, after

which it is first passed through that part of the coil which is closest to the radiant heat source. Upon entry into the coil, the feed is still substantially liquid, but as cracking proceeds, gas is formed and the flow through the coil changes from liquid to gas/liquid. In order to allow the feed to be adequately heated, the velocity of the feed through the coil should be chosen such that an optimum flow regime through the entire coil is obtained. It has been found particularly advantageous that the liquid hydrocarbon oil feed enters the cracking furnace at the feed inlet with a velocity in the range of from 1 to 6 m/s and the cracked product leaves the furnace at such velocity that an annular flow of gas and liquid is obtained at the outlet. In practice, it has been found that this implies that the velocity of the gas/liquid mixture at the outlet of the coil should be in the range of from 50 to 120 m/s, preferably from 55 to 100 m/s and more preferably from 60 to 80 m/s.

An annular flow refers to the situation wherein liquid flows as a ring along the walls of the tube and covers the entire wall, whilst the gas flows through the centre. If the velocity of the gas/liquid mixture in the coil would be too high, unacceptable pressure drop across the coil would occur together with undesired flow patterns. In case of too high a velocity of the gas/liquid mixture mist flow occurs inside the coil, i.e. a situation wherein the coil is entirely filled with vapour with some wetting at the walls of the tube. Too low a velocity of the gas/liquid mixture will cause slug flow in vertical tubes and stratified flow in horizontal tubes. Slug flow refers to the situation wherein the gas/liquid mixtures flows through the tube as successive packets of liquid and packets of gas, whilst stratified flow of a gas/liquid mixture means that liquid flows along the bottom part of the tube with the gas flowing

above this liquid. It will be evident that an annular flow is the optimum situation for a uniform heating of the liquid in the coil via transfer of radiation heat through the tube wall.

Figure 1 shows a top view of the radiant section of a cracking furnace having a coil lay-out in accordance with the present invention.

Figure 2a is a top view of a preferred radiant section of a cracking furnace according to the present invention.

Figure 2b is a side view of the sinus-shaped coil applied in the radiant section depicted in figure 2a.

Figure 3 is a schematic top view of a cracking furnace comprising four integrated radiant sections.

Figure 4 is a schematic top view of a conventional furnace with separate heating and soaking sections.

In figure 1 the liquid hydrocarbon oil feed enters the radiant section (7) at point (1), where it is first passed to the inner row (2) in which it is heated by heat from radiant heat source (6) to such temperature that cracking occurs. Prior to entry in the radiant section the feed may have been passed through a convection section (not shown) for preheating purposes. At the end of the inner row (2), the (partially cracked) feed flows to the outer row (4) at point (3), where further cracking takes place. At the end of the outer row (4), the cracked product leaves the radiant section (7) at point (5).

In figure 2a the same procedure as indicated in figure 1 is followed whereby the radiant section contains four burners (6) and the coil has a sinusoid-shape as indicated in figure 2b.

Figure 3 schematically shows the embodiment in which four radiant sections are combined into a single containment. The reference numbers have the same meaning as in figure 1. The inner rows of the individual radiant

sections are combined in a single inner row (2). The single outer row (4) surrounds the four radiant heat sources (6) and the single inner row (2).

In Figure 4 a conventional cracking furnace is schematically depicted. Three separate coils indicated by postscripts a, b and c enter the heating section (7) of the cracking furnace at points (1a), (1b) and (1c), respectively. In heating parts (2a), (2b) and (2c) of the coils cracking reactions are initiated and proceed to a certain conversion level. Via connections (3a), (3b) and (3c), the coils proceed in soaking section (8) in soaking parts (4a), (4b) and (4c). The coils finally leave the soaking section (8) at points (5a), (5b) and (5c). Both heating section (7) and soaking section (8) contain a radiant heat source (6).

The invention is further illustrated by the following example without restricting the scope of the present invention to this particular embodiment.

<u>Example 1</u>

A deasphalted Arab Heavy long residue (AH LR) is fed into a vertical cracking furnace containing one radiant section as indicated in figure 2a, i.e. having a square horizontal cross-section with four burners in the centre. The coil is made of a conventionally applied alloy comprising chromium and molybdenum (Cr/Mo weight ratio of about 10/1). The furnace has a design throughput of 3,000 tonnes of feed per day. Further characteristics of the radiant section are:

(1) an inner lining of 8.8 m,

(2) a height of 16.3 m,

(3) a single coil arranged in two square rows in a sinusoidal mode as depicted in Figure 2b, whereby:

  (a) the inner coil row has a diameter of 6 inch (15 cm) and has 64 points of intersection with the horizontal cross-sectional plane,

(b) the outer coil row has 100 points of intersection with the horizontal cross-sectional plane, and

(c) up to and including the first 76 points of intersection, the outer coil row has a diameter of 6 inch, after which the diameter expands to 8 inch (20 cm).

Further details with respect to the operating conditions and the results of the thermal conversion are indicated in Table I in the column denoted as E-1.

Comparative Example 1

A deasphalted Arab Heavy long residue (AH LR) is fed into a conventional coil cracking furnace consisting of a vertically arranged heating section and a vertically arranged soaking section and three coils each having a diameter of 4 inch (10 cm) and a design throughput of 1,000 tonnes of feed per day. Each section has an inner lining of 8.9 m, a height of 16.3 m, round horizontal cross-section with three burners in the centre. The coils are made of the same material as used in Example 1. Each coil has 23 points of intersection with the horizontal cross-sectional plane in the heating section and also 23 points of intersection with the horizontal cross-sectional plane in the soaking section. A top view of the furnace is schematically indicated in Figure 4.

Further details with respect to the operating conditions and the results of the thermal conversion are indicated in Table I in the column denoted as CE-1.

Example 2

Example 1 is repeated, except that a furnace is used having a single coil made of an alloy comprising nickel and chromium in a weight ratio Ni/Cr of about 1.4/1.

Further details with respect to the operating conditions and the results of the thermal conversion are indicated in Table I in the column denoted as E-2.

TABLE I Thermal conversion of deasphalted AH LR

	E-1	CE-1	E-2
Furnace inlet temperature (°C)	370	370	370
Furnace outlet temperature (°C)	460	505	460
Coil skin temperature SOR[1] (°C)	550	550	550
Coil skin temperature EOR[2] (°C)	650	650	950
Outlet pressure SOR (bar)	13	13	13
Run length (hours)	6100	1600	18300
520 °C+ conversion (%wt)	77	77	77

[1] start of run

[2] end of run

Table I shows that the furnace used in Example 1 (E-1) can be in continuous operation for 6100 hours, thereby converting 77% by weight of the material having a boiling point of 520 °C or higher present in the feed to lower boiling material. After this period of 6100 hours the furnace has to be taken out of operation for decoking of the inner coil row. The conventional furnace, however, has to be taken out of operation already after 1600 hours of operation for decoking of the coils in the heating section. The furnace with the coil made of high coking resistant material, finally, can be in continuous operation for 18300 hours before decoking has to take place. This long period of operation is the result of the much higher coil skin temperatures that can be reached before the coke deposition onto the coil skin reaches such level that decoking is necessary.

From Table I it can be seen that when using the same coil material, the furnace having the configuration of according to the present invention (E-1) can be operated much longer at comparable conditions than when using a conventional furnace having the same capacity in terms of throughput (CE-1).

CLAIMS

1. Cracking furnace comprising a containment having a feed inlet and an outlet for cracked product, in which containment is arranged at least one radiant section comprising a coil and a radiant heat source, characterised in that a single coil is arranged around the radiant heat source in at least two co-axial rows, whereby the opening of the coil at that part of the coil forming the innermost row is connected to the feed inlet and the opening of the coil at that part of the coil forming the outermost row is connected to the outlet for the cracked product.

2. Cracking furnace according to claim 1, wherein the single coil is arranged in two rows around the radiant heat source.

3. Cracking furnace according to claim 1 or 2, wherein the single coil is made of an alloy comprising chromium and molybdenum or, preferably of an alloy comprising nickel and chromium.

4. Cracking furnace according to any one of claims 1 to 3, wherein the diameter of the coil at the feed inlet is smaller than the diameter of the coil at the outlet for the cracked product.

5. Cracking furnace according to claim 4, wherein the diameter of the coil is the same throughout the innermost row.

6. Cracking furnace according to any one of the preceding claims, wherein the radiant heat source comprises at least one burner, preferably from three to six burners.

7. Cracking furnace according to any one of the preceding claims, wherein the cracking furnace is a vertical furnace with the radiant section having a substantially square horizontal cross-section, whereby

the single coil is arranged in two substantially square rows around the radiant heat source.

8. Process for the thermal conversion of a hydrocarbon oil feed wherein use is made of a cracking furnace according to any one of claims 1 to 7.

9. Process according to claim 8, wherein the hydrocarbon oil feed enters the cracking furnace at the feed inlet with a velocity in the range of from 1 to 4 m/s and the cracked product leaves the furnace at such velocity that an annular flow of gas and liquid is obtained at the outlet.

10. Process according to claim 8 or 9, wherein the hydrocarbon oil feed is an atmospheric residue, a vacuum residue, a deasphalted oil or a mixture of two or more of these.

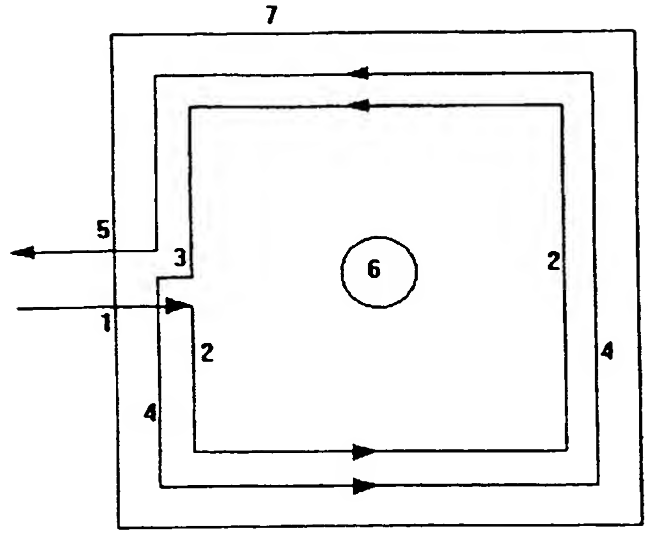

Figure 1

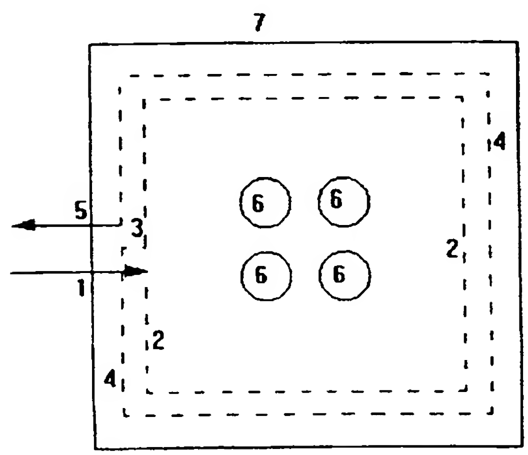

Figure 2a

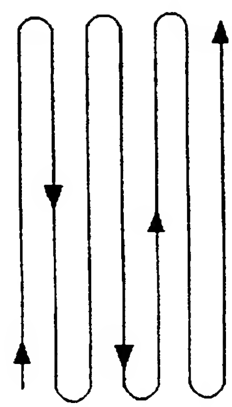

Figure 2b

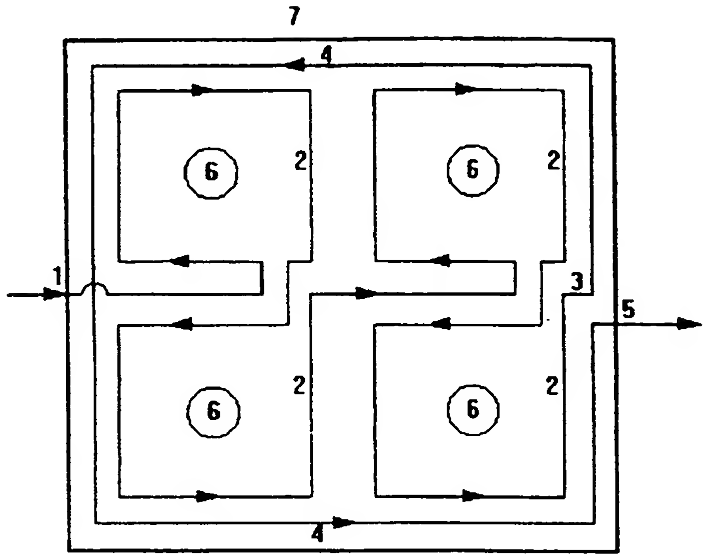

Figure 3

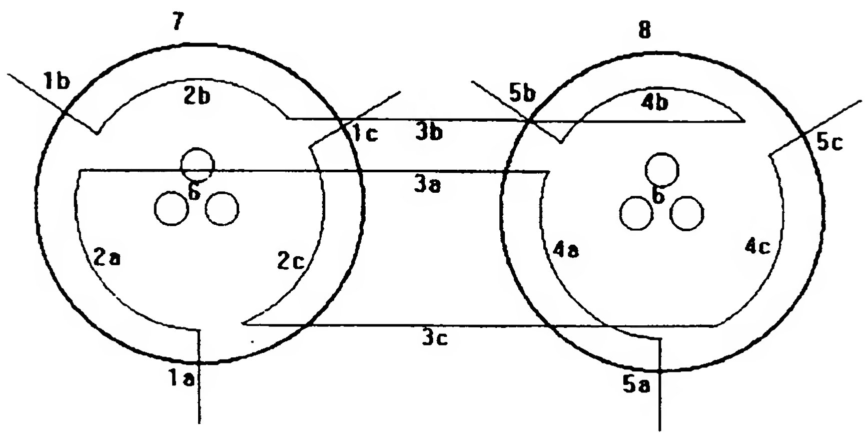

Figure 4

INTERNATIONAL SEARCH REPORT

International Application No
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A. CLASSIFICATION OF SUBJECT MATTER
IPC 6 C10G9/20

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 6 C10G

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PETROLEUM REFINER, vol. 29, no. 1, January 1950, HOUSTON, pages 109-118, XP002029496 CAMPBELL: "Cylindrical furnaces for the petroleum industry" see figures 7,9 ---	1-10
A	US 2 182 586 A (UOP) 5 December 1939 see figures 1,2 ---	1-10
A	US 2 246 469 A (GASOLINE PRODUCT COMPANY) 17 June 1941 see figures 1,2 ---	1-10
A	FR 1 325 244 A (SHELL) 26 July 1963 ---	
A	US 3 512 506 A (VON WIESENTHAL) 19 May 1970 -----	

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents :

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Date of the actual completion of the international search	Date of mailing of the international search report
15 April 1997	24.04.97

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL - 2280 HV Rijswijk Tel. (+ 31-70) 340-2040, Tx. 31 651 epo nl, Fax (+ 31-70) 340-3016	Michiels, P

INTERNATIONAL SEARCH REPORT

Information on patent family members

Int ional Application No
PCT/EP 97/00451

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
US 2182586 A	05-12-39	NONE	
US 2246469 A	17-06-41	NONE	
FR 1325244 A	26-07-63	NONE	
US 3512506 A	19-05-70	NONE	